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THE GREEN AREA EFFECT AND OTHER AEROSOL SCAVENGING PROCESSES IN THE BLACK HILLS OF SOUTH DAKOTA: FINAL REPORT

By: Steven J. Haggard

Prepared for:

Rocky Mountain Forest and Range
Experiment Station
Fort Collins, Colorado 80521

Contract Nos. 16-419-CA & 16-538-CA

Institute of Atmospheric Sciences

South Dakota School of Mines and Technology

Rapid City, South Dakota 57701

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ABSTRACT

Bi-weekly air sampling for particulates in the Black Hills has revealed several important scavenging mechanisms. A Gardner condensation nucleus counter was used to determine these particulate counts at fourteen stations. Analysis of the data has indicated that the forested Black Hills are a source of clean air when compared to the surrounding plains due mainly to (a) greater precipitation and in-cloud scavenging events, (b) an elevation variation, and (c) the filtering effects of vegetation (green area sink effect). Little or no scavenging was indicated by certain meteorological variables when considered separately. The green area sink effect was substantiated after performing several tests on the data with a cleanup of as much as 50% evidenced.

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1. INTRODUCTION

1.1 Background on Atmospheric Aerosols

Aerosols (particulates, nuclei) are minute particles ranging in size from .005 to 500 microns (μm). They can be as small as a cluster of several molecules or as large as a visible dust kernel. In the atmosphere, they are usually classified by size. Particles larger than 5 μm in radius are affected by gravitational settling and are classified as "dustfall." Smaller particles tend to remain airborne for indefinite periods of time and are classified as "suspended particulates." Of these, the smallest ones behave like a gas or vapor and are highly subject to Brownian motion, which is motion imparted by collisions with air molecules. A further classification by size (radius) is as follows: $r<0.1$ μm are called Aitken nuclei, $0.1<r<1.0$ μm are called large nuclei, and $r>1.0$ μm are called giant nuclei.

The maximum concentration of particulates occurs in the .01 to .1 μm radius range and falls off rapidly as the radius increases (Byers, 1965). These particles are referred to as "Aitken particles" after John Aitken, who designed one of the first expansion chambers to count nuclei. It was originally believed that hygroscopic Aitken particles were active as cloud condensation nuclei, but it is now assumed that larger hygroscopic particles (.1 to 3 μm) are activated more readily in nature, since they require smaller supersaturations than Aitken particles.

Some of the common sources of aerosols are as follows: (1) dust from wind erosion, (2) natural and man-made combustion, (3) sea spray, (4) volcanic eruptions, (5) meteoric dust, (6) chemical and photochemical reactions, and (7) organic sources (pollen, terpenes). In Appendix A, the sources most prevalent in the Black Hills regions will be discussed.

1.2 Purpose of Study

The data collected in this study have been analyzed to evaluate the effects of scavenging mechanisms for aerosols in the Black Hills region. One such mechanism, the "green area" sink effect, is the focal point of this study.

1.3 The Green Area Sink Effect

This particular scavenging mechanism is based on the hypothesis that an area of high density vegetation can act as an air filter, removing aerosols by dry deposition. In this case, the Black Hills region with its high density pine-spruce-aspen forests, is hypothesized to take on the characteristics of a "green area."

The "green area" sink hypothesis is supported by the results of previous studies. In an early comprehensive study of condensation nuclei, Landsberg (1938) mentioned measurements of nuclei concentrations obtained in various German cities in the 1930's. In Wiesbaden, a city with many parks, a comparison was made between counts in the business district and counts in the park areas. Results showed an average of 100,000 nuclei cm^{-3} in the business district and 35,000 nuclei cm^{-3} in the parks. Neuberger (1948), another of the early investigators, also recognized the importance of vegetation as an aerosol filter in his study of nuclei.

More recent systematic studies show additional support for the green area sink effect. Hyde Park in London, England (Meetham, 1964) showed a 27% reduction in smoke concentration when compared to surrounding areas of the city. A similar effect was demonstrated in Cincinnati, Ohio (Bach, 1971) in a park-like cemetery near an industrial complex. Along a New Jersey highway (Motto, 1970), the lead content in roadside plants was shown to decrease at increasing distances within 100 feet of the roadway.

Trees seem to be the most effective scavengers of particulates from the atmosphere when compared to other plants. Smith (1974) indicated that this is due to high surface area to volume ratio of their foliage. Neuberger (1967) found that coniferous trees were more efficient than deciduous trees in the scavenging of suspended particulates. An eight-month study was conducted near Steubenville, Ohio (Dochinger, 1969) to measure dustfall and suspended particulates at two sites with three stations each. One station per site was located in open terrain, one under a hardwood canopy, and one under a coniferous canopy. Hi-volume samplers were used to measure suspended particulates over 24-hour periods, and dustfall buckets were used for measurements of dust. The results showed that coniferous trees were most effective in the removal of suspended particulates as well as dusts.

The theory behind the green area sink effect and the processes involved have also been explored. Ricks and Williams (1974) concluded that gases are removed from the atmosphere by a diffusion process primarily through the stomatal openings on the leaf surfaces. They also concluded that particulates are collected or adsorbed on the leaf surfaces. Smith (1974) developed several classes of pollution-ecosystem interactions and concluded that particulates may be scavenged from the atmosphere via impaction on plant surfaces. Aylor (1975) conducted experiments involving ragweed pollen (r=20 μm) deposition in a corn field. He concluded that stronger winds resulted in greater scavenging of pollen by the corn canopy due to the increase in impaction with increased wind speed. Motto (1970) indicated that the lead particles collected by roadside plants may be present as surface adsorbed material or may be passed

into the plant through the stomatal openings due to the small particle size of airborne lead.

1.4 Collection of the Black Hills Data

For the past two years, the Institute of Atmospheric Sciences (IAS) has been supported by the Rapid City unit of the Rocky Mountain Forest and Range Experiment Station, U.S. Department of Agriculture, in the collection and analysis of the data used in this study. The monies involved were allotted by the SEAM program, which is a program designed to research, develop, and apply technology to maintain environmental quality while helping meet the nation's growing energy requirements.

The air quality measurements obtained in this project are expected to provide a reasonable baseline particulate count for this area. This is of particular interest at this point in time because of the anticipated development of the coal reserves to the west and northwest of the Black Hills, which is expected to alter this baseline count. According to Bryson (1972), there is increasing evidence that many desert conditions in the world were originally caused by an increased particulate loading in the atmosphere, which resulted in over-competition among condensation nuclei for the available water vapor and the creation of inefficient clouds. The Thar Desert of India and Pakistan, which was once a productive agricultural region, was cited as an example. In that case, the increase in particulates was caused by overgrazing followed by wind erosion.

Figure 1 shows the locations of the 14 sampling sites in and around the Black Hills area of South Dakota and Wyoming. In order to develop a data comparison, two sets of seven stations each were established. The two sets are referred to as "plains stations" and "hills stations;" the

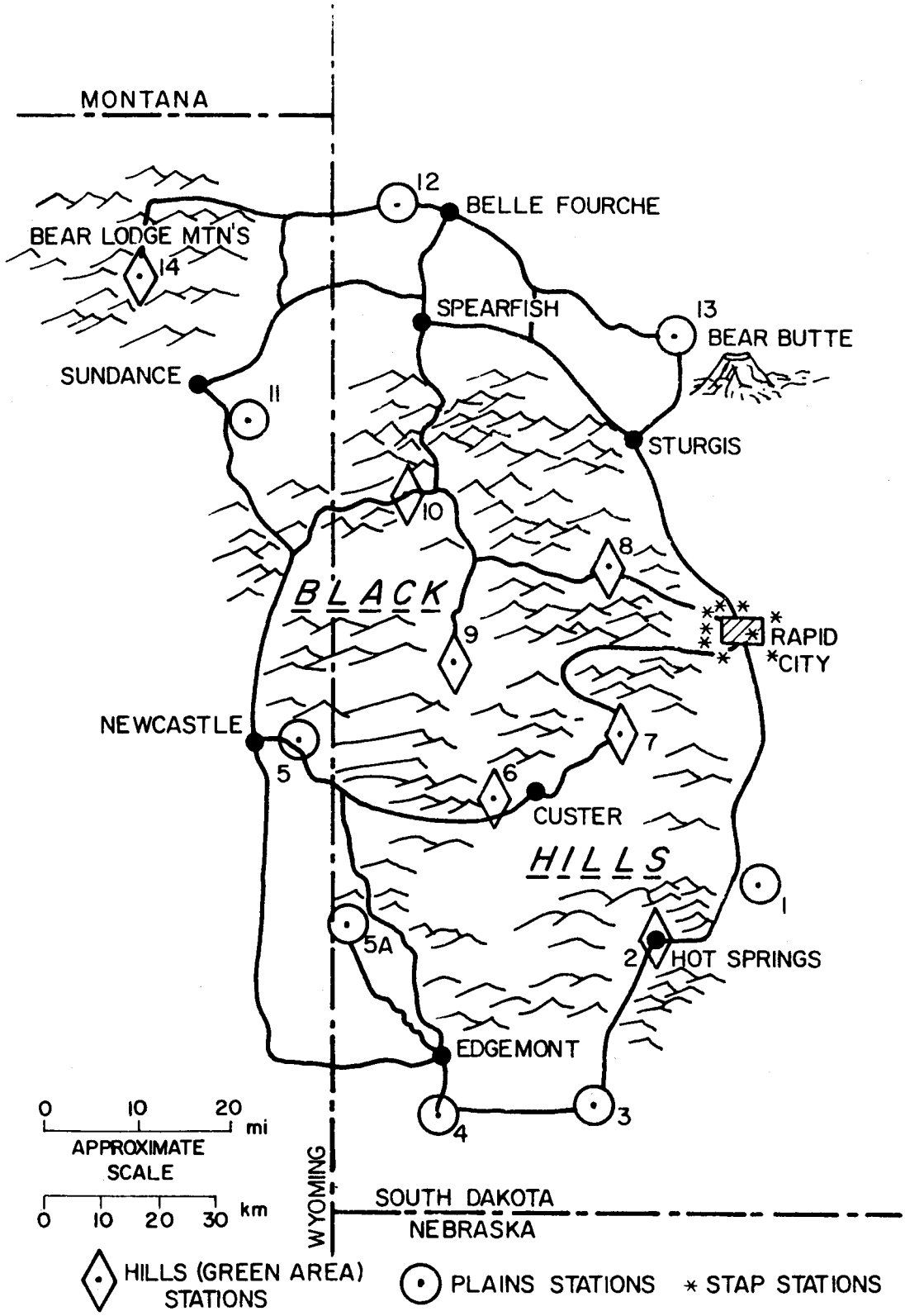


Fig. 1. Air sampling circuit showing locations of individual stations in and surrounding the Black Hills.

former being located on open prairie or farmland, and the later in the forested green area.

The coverage of the 14 sites was divided into two circuits; a southern circuit covering Stations 1-7, and a northern circuit covering Stations 8-14. Sampling runs were conducted every second week on the middle days of the week (typically, Wednesday and Thursday), with the southern circuit traversed on the first day and the northern circuit on the following day. In addition, the sampling order of the stations was reversed each time a circuit was covered. Sampling runs were conducted from May 28, 1974 to May 6, 1976 for a total of 50 samples per station.

Each time a sampling site was visited for the purpose of data collection, meteorological conditions were recorded as well as particulate counts. Table 1 is a sample of a completed field station report. Wind direction and speed were measured with a portable wind vane and a hand-held anemometer. Dry- and wet-bulb temperatures (from which relative humidity was calculated) were obtained with a sling psychrometer, and aerosol concentration was determined with a Gardner condensation nucleus counter.

The Gardner counter used is pictured in Fig. 2. It has been modified from the original model by the addition of a more precise vacuum gage in order to measure nuclei counts at small as well as large pressure differentials.

The following procedures are involved in the operation of the Gardner counter:

(1) An air sample is pulled into a humidifying chamber and allowed to rest while an adjacent chamber is evacuated to a desired pressure.

TABLE 1

Cloud Physics Laboratory — Institute of Atmospheric Sciences

Field Station Report

Date 5-5-76 Observer DB Station 7

Start Time 0900

End Time 0915

A. Meteorological Observations

1. Wind Direction 340 deg. 2. Wind Speed 3 mph

3. Dry Bulb T. ____ °C 39 °F 4. Wet Bulb T. ____ °C 38 °F

5. Est. Percent Cloud Cover 100.

6. Cloud Type (circle) ____ Est. Height Above Station 200 ft.

a. Cirrus b. Altostratus c. Altocumulus
(d.) Stratus e. Stratocumulus f. Nimbostratus
g. Cumulus h. Cumulonimbus i. Fog

7. Precipitation Class: Observed Rate light (in) ____ (mm)

(a.) $P < 2.5$ mm hr^{-1} b. $2.5 \leq P < 7.5$ mm hr^{-1} c. $P \geq 7.5$ mm hr^{-1}

8. Precipitation Type

(a.) Drizzle b. Rain c. Snow d. Hail e. Ice crystals

9. Est. Percent snow cover (over area with 2 mi-radius or station) 5, average snow depth ____.

10. Est. time of last precipitation at station. (Hours previous to visit up to 48) 1.

11. Est. visibility (miles) 2.

12. Other observations (e.g., blowing dust, heavy pollen production, etc.)

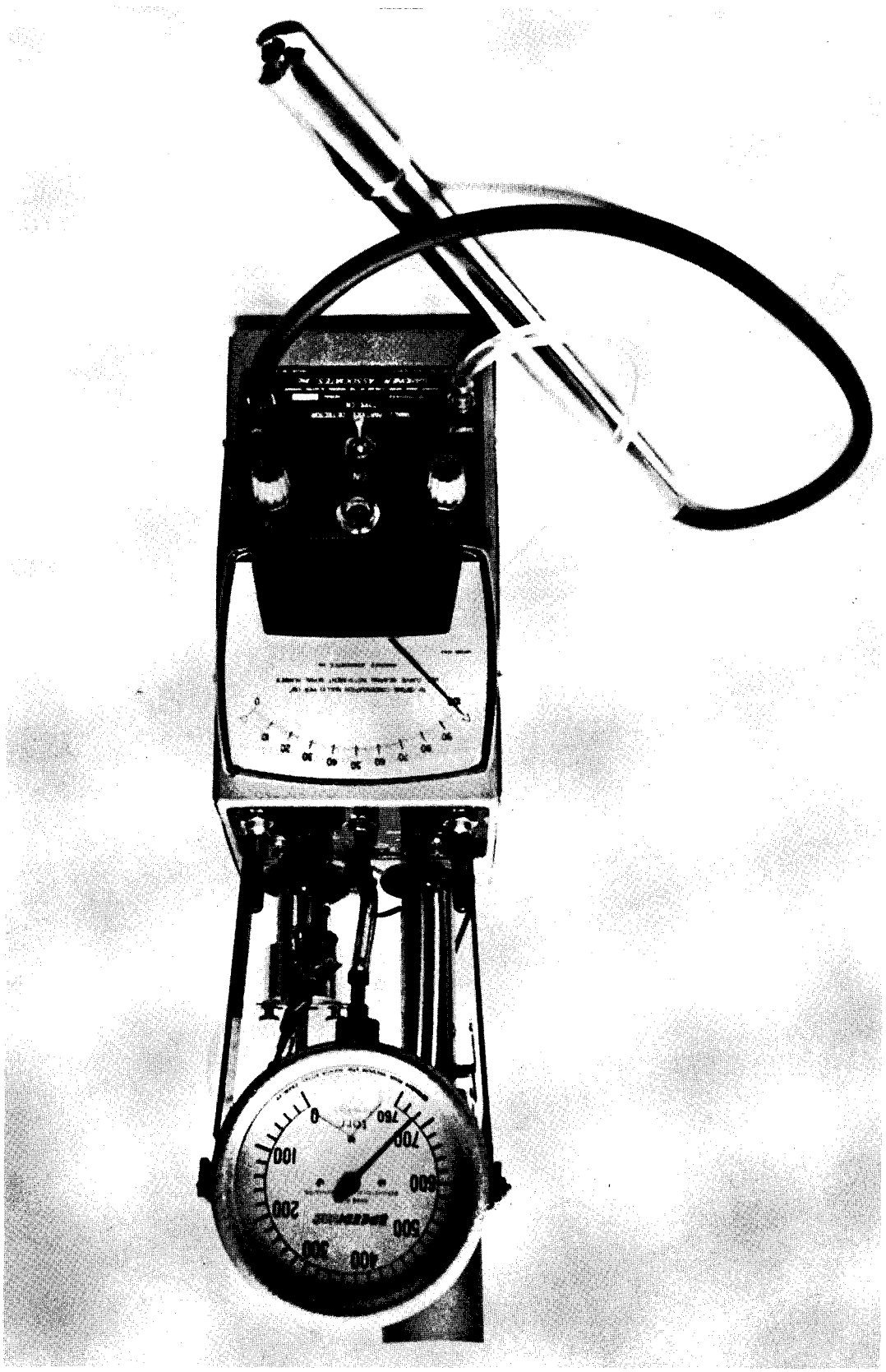
fog covering tops of peaks

B. Condensation Nuclei Count (expansion type)

1. Instrument C.N-2 2. Vacuum setting a. MAX VAC-

3. Vacuum setting b. 10 torr 4. Vacuum setting c. ____

5. Count a. 6, 7, 6 6. Count b. 2, 2 7. Count c. ____



(2) The humidified air is then allowed to expand into the evacuated chamber. This rapid adiabatic expansion produces a cloud, the density of which is proportional to the number of particles activated as condensation nuclei.

(3) A photocell measures the opacity of the cloud and transfers this measurement to a voltmeter.

(4) The voltmeter reading is then compared to a calibration curve to determine the actual nuclei concentration.

At an approximate minimum pressure differential of 10 torr (1 torr= 1 mm Hg), a nominal saturation ratio of 1.04 is produced, activating true cloud condensation nuclei (CCN) plus some larger Aitken nuclei. Davis (1975) has referred to these as "near-CCN" particles. At the maximum pressure differential (600-650 torr), saturation ratios greater than 3.0 are produced, thus activating most all the nuclei present, resulting in an "Aitken count."

2. ANALYSIS OF THE DATA

2.1 General Methods

In order to simplify most of the data analysis, all information from the field station reports was put onto data cards which could then be used in conjunction with various computer programs. This proved to be a highly efficient method of minimizing analysis time and the human error factor.

In some cases it was necessary to determine a mean of nuclei counts for individual stations, or for a combination of stations such as all the hills stations or all the plains stations. In such cases, the geometric mean was used in preference to the arithmetic mean because of its ability to suppress extreme values. The geometric mean of counts, designated $\hat{C}$ hereafter, is defined as follows:

$$\hat{C} = \left(\prod_{i=1}^{n} C_i\right)^{\frac{1}{n}} ,$$

i.e., the n^{th} root of the product of n observations.

Standard statistical methods were applied to the quantitative data from the field station reports. Linear and multiple linear regressions were the most commonly used devices, since it was often necessary to show whether or not correlation existed between nuclei counts and certain independent variables. Correlation coefficients thus obtained were then tested against tabulated maximum values that could have been obtained randomly, in order to determine whether the correlation was real or whether it was coincidental. If a correlation coefficient obtained was greater than a tabulated value for the same sample size at 95% confidence,

this would say that 95 times out of 100 the correlation between the variables would be real, and 5 times out of 100 it would be coincidental.

When comparisons were made between data groups with normal distributions to determine whether or not they came from the same population, a test statistic was used. In cases where the normality of a distribution was not established, the non-parametric test was used. The only one used herein was the Wilcoxon-Mann-Whitney (rank-sum) test.

The original Station 5, located near Newcastle, Wyoming (refer to Fig. 1), was omitted from all data analysis as a statistical outlier. Aitken nuclei counts at that site were consistently high, and it was concluded that its location was such that the plume from the city of Newcastle often contaminated it. So Station 5 was relocated (as of the 25^{th} sampling run) to a point just south of Dewey, South Dakota (Station 5A, Fig. 1).

The Grubb's test was the statistical device used to determine S-5's status as an outlier. Its application to this problem follows. First of all, the geometric means of Aitken counts at each of the plains stations as of the 24^{th} sampling run were arranged in ascending order. Next, the mean of these observations was determined, first with all seven points included, then with the suspect point (S-5) omitted. Following this, the squares of the deviations from the two means were determined, first with all seven points included, then with the suspect point omitted. The ratio of the two squares of deviations was calculated, then compared to tabulated values to determine at what significance level the suspect observation could be rejected as an outlier. In this case, the significance level was .05, which for these data demonstrate good justification for throwing out the observations of Station 5 up to the 25^{th} sampling.

3. RESULTS OF ANALYSIS

3.1 General Observations

Figures 3a and 3b show geometric mean Aitken counts ($\hat{C}_{max}$) and geometric mean "near-CCN" counts ($\hat{C}_{10}$) obtained for hills and plains stations for each sampling run from May 28, 1974 through May 6, 1976. There is a large variation from sample to sample which is primarily due to the different types of weather systems that pass through the area.

Figures 4 and 5 show contours of geometric mean Aitken and near-CCN counts respectively for the entire sampling area. These figures show that the lowest particulate concentrations are centered within the Black Hills, especially the northern hills. There are several explanations for this phenomenon; a natural decrease in counts with increasing elevation, a decrease due to precipitation scavenging which occurs more strongly and more frequently in the hills than over the surrounding plains, and the green area sink effect. Each of these will be discussed individually in later sections.

3.2 Effects of Precipitation and In-Cloud Scavenging

Clouds and precipitation are well-known scavengers of suspended particulates. Nucleation processes and subsequent collection between cloud droplets and aerosols remove a portion of these tiny particles, but the washout process by precipitation is much more efficient as a scavenger. According to Byers (1965), raindrops are more likely to capture larger aerosol particles, with greater washout occurring with rains of stronger intensity and longer duration. Snowflakes tend to be more effective as scavengers for the smaller aerosols.

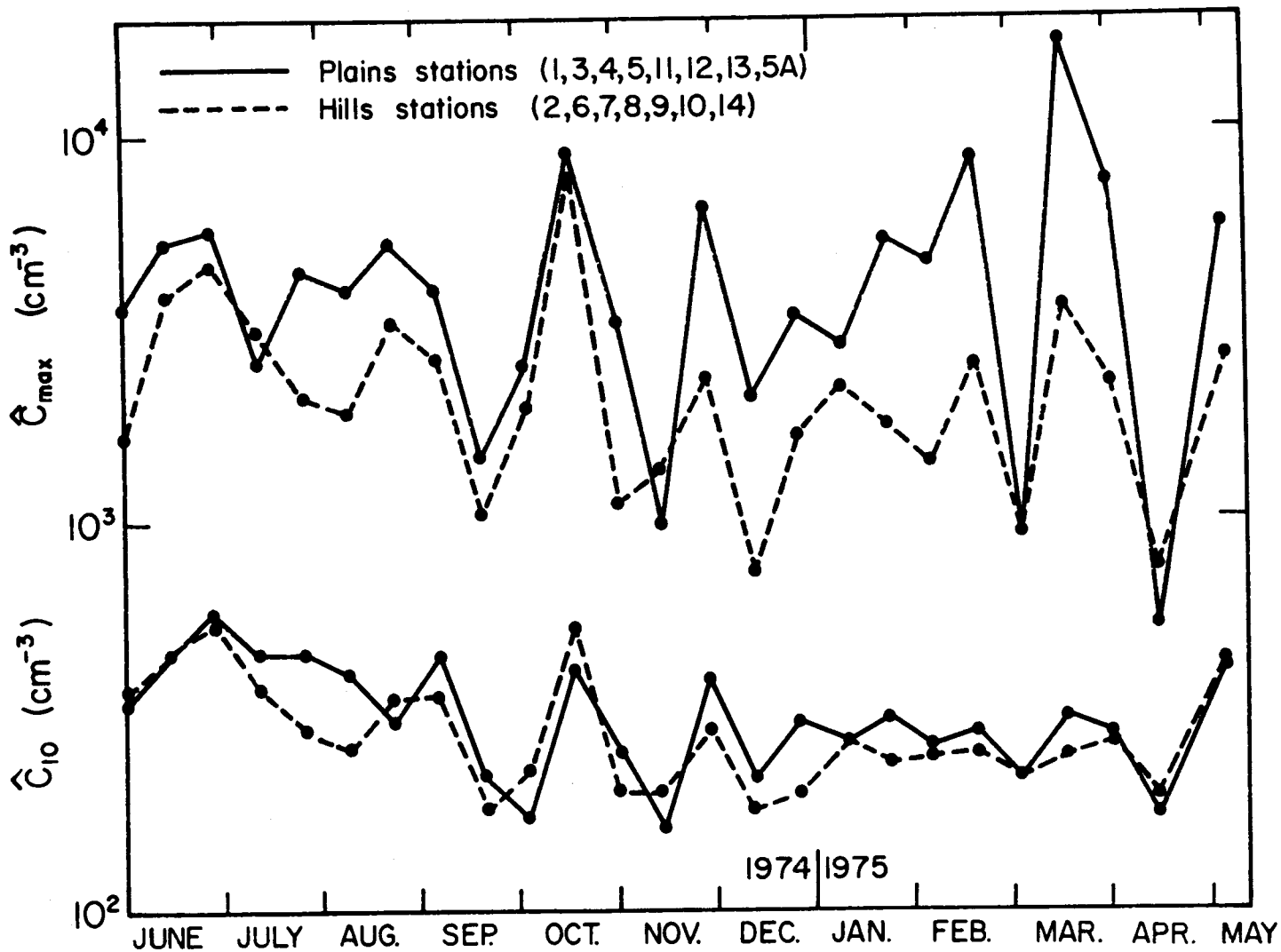


Fig. 3a. Geometric mean Aitken and near-CCN nuclei counts as a function of time for first-year sampling period.

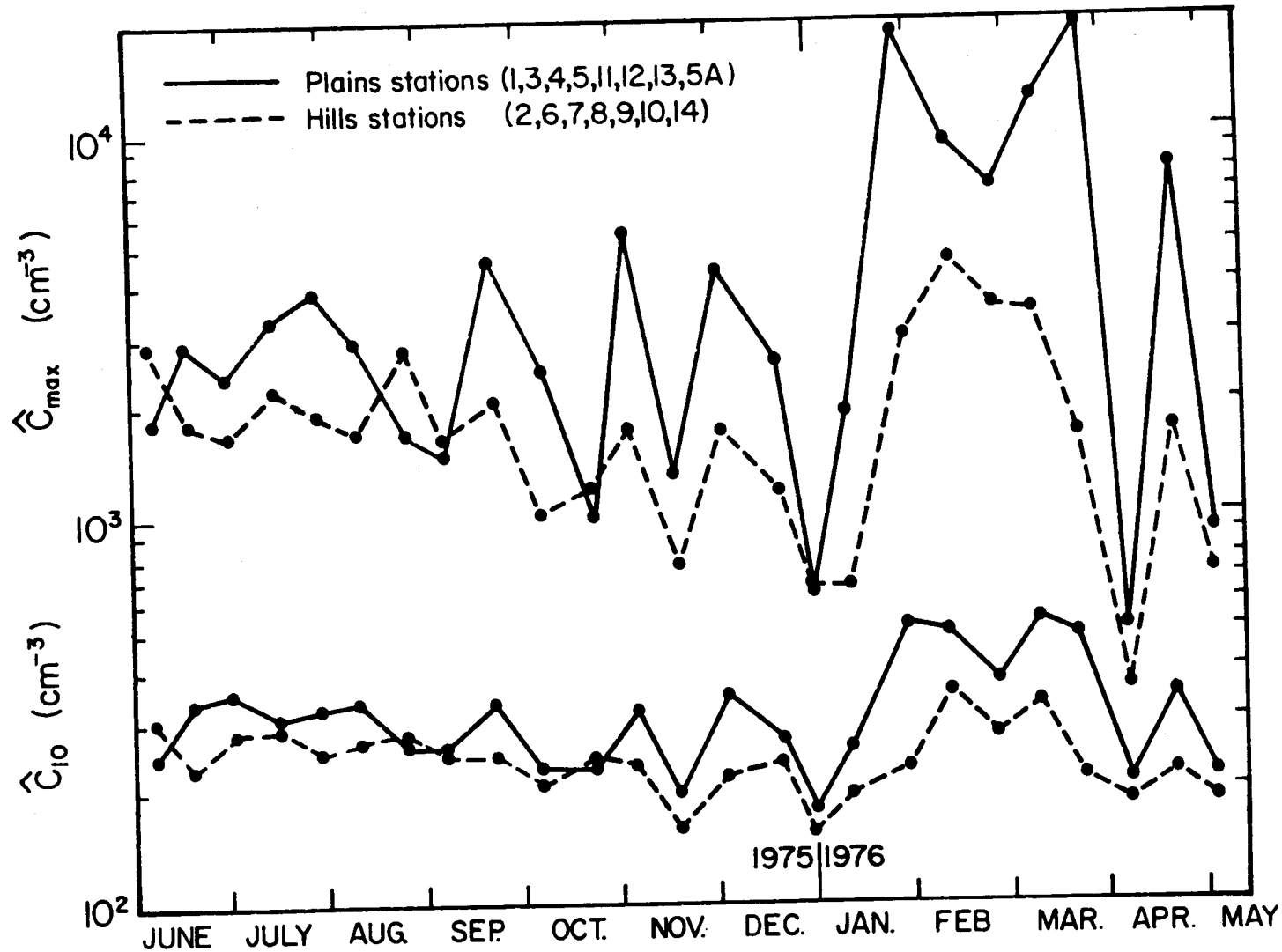


Fig. 3b. Geometric mean Aitken and near-CCN nuclei counts as a function of time for second-year sampling period.

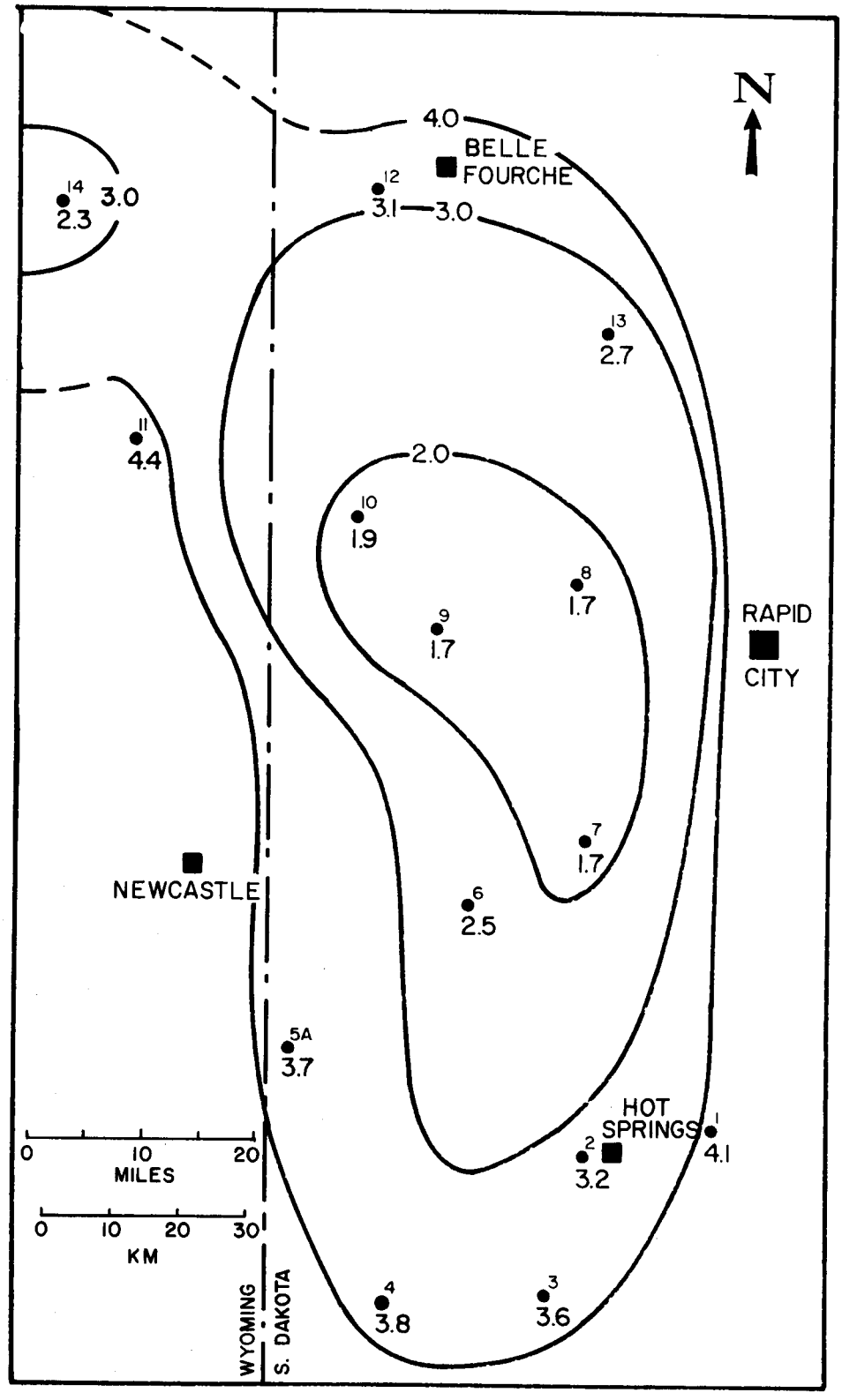


Fig. 4. Isopleths of Aitken nuclei concentration in units of 10^3 cm^{-3} for 14 sampling locations. Station number lies to the upper right and concentration immediately below station location.

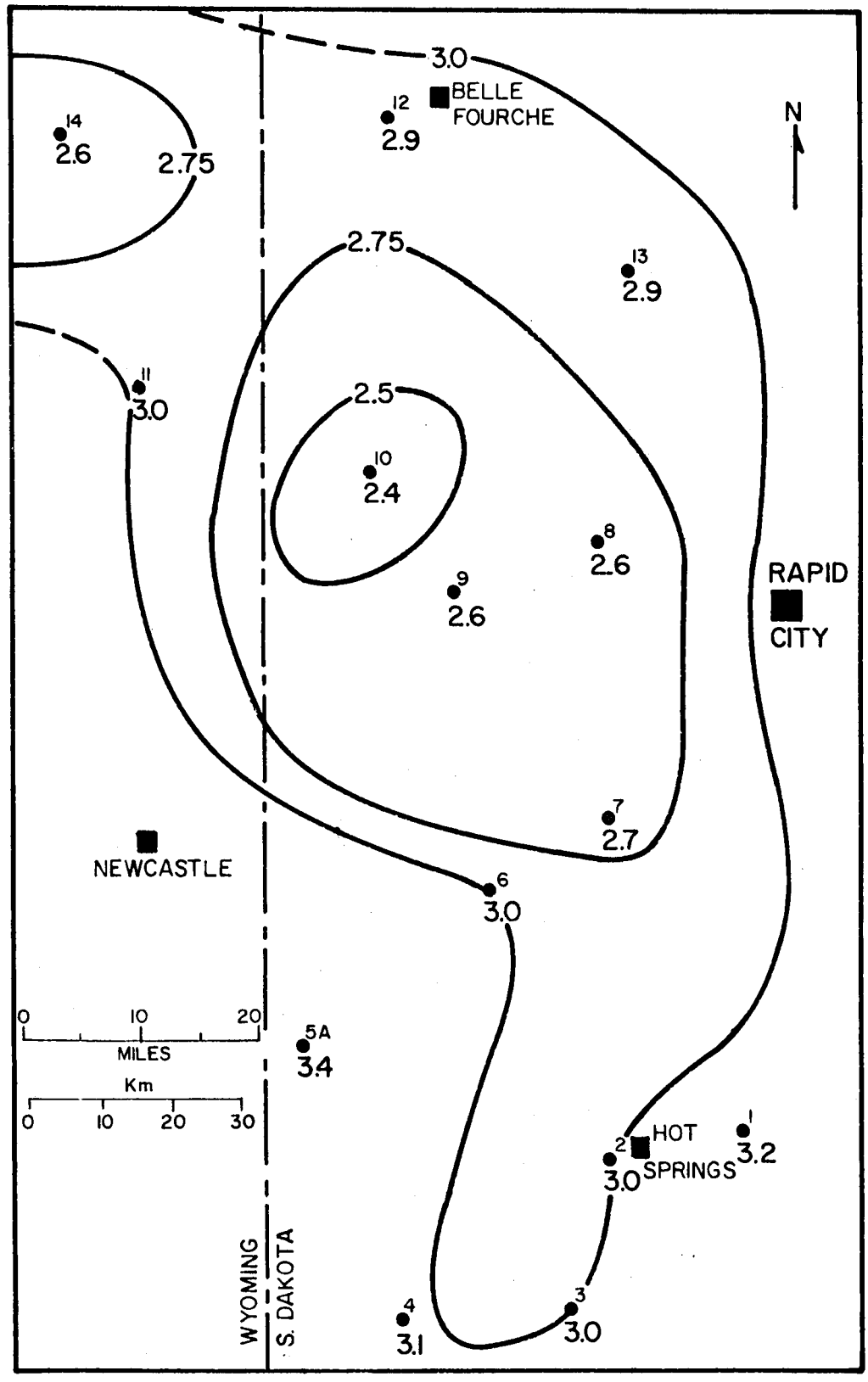


Fig. 5. Isopleths of near-CCN nuclei concentration in units of 10^2 cm^{-3} for 14 sampling locations. Significance of numbers as in Fig. 4.

Table 2 shows the effects of precipitation scavenging as evidenced by the data. Geometric mean Aitken counts are compared for non-precipitation observations, rain/drizzle observations, and snow observations for hills and plains stations. The cleanup is quite evident for both liquid and frozen precipitation, with a slightly greater scavenging effect for snow than for rain. Note that there is little difference between hills and plains stations when precipitation is reported. Because of this "equalizing" effect between hills and plains stations, all precipitation scavenging observations have been omitted from statistical analyses and green area tests.

TABLE 2

Effects of Precipitation Scavenging

Plains			Hills		
Type of Observation	$\hat{C}_{max}$	n	Type of Observation	$\hat{C}_{max}$	n
Dry	3563	302	Dry	2095	283
Rain/Driz.	785	9	Rain/Driz.	712	11
Snow	406	15	Snow	426	28

3.3 Effects of Specific Synoptic Weather Systems

Weather systems on a synoptic scale are known to have an effect on local pollutant concentrations. One only need look at the Thanksgiving, 1966, pollution episode on the east coast of the U.S. (Air Pollution Control Administration, 1968). In that case, a dynamic anticyclone stagnated over the eastern part of the country. The strong subsidence inversion which developed limited vertical dispersion of the pollutants. The episode lasted for several days and was finally broken by the passage of a cold front.

Western South Dakota is occasionally influenced by warm, semi-stationary anticyclones during the fall and winter months. These so-called "Great Basin highs" persist over the western U.S. between the Rocky Mountains and the coastal ranges for several days to over a week before dissipating. Vertical dispersion of pollutants in this region should be limited due to the subsidence inversions resulting from these persistent highs. Two strong cases of presence of high particulate concentrations while under the influence of Great Basin highs are evidenced in the data. Both cases occurred during the first two months of 1976.

Case 1: January 28-29, 1976, sampling run #43. A Great Basin high had developed on January 25 over northeast Nevada, reaching a maximum central pressure of 1040 millibars (mb) over western Colorado on January 27. Ridging of both contours and isotherms at 500 mb was very strong in this case. During the sampling runs of January 28-29, the central pressure of the surface high was about 1034 mb over northeast Utah with an anticyclonic circulation affecting a large portion of the western U.S. The Rapid City soundings showed early morning subsidence inversions near 700 mb on both days, which lowered to 760 mb by the afternoon of January 29. Geometric mean Aitken counts for plains and hills stations were as follows: $\hat{C}_{max}$(plains)=16,830 cm^{-3} and $\hat{C}_{max}$(hills)= 3426 cm^{-3}. Two hills stations, S-8 and S-9, visited on the morning of January 29, reported very low counts which accounts for the low hills mean. All other hills stations reported higher than normal counts. Upon examination of the soundings and the meteorological data from the field station reports, no plausible explanation could be arrived at which would account for these low readings at S-8 and S-9.

Case 2: February 11-12, 1976, sampling run #44. A Great Basin high had developed over southwest Idaho on February 10, reaching a maximum central pressure of 1036 mb on February 11 over northeast Nevada. The ridging aloft in this case was very weak and short-lived. During the sampling runs of February 11-12, the central pressure of the surface high decreased from the maximum of 1036 mb to 1022 mb by the afternoon of February 12, with the circulation not being as widespread as in Case 1. Geometric mean Aitken counts for plains and hills stations were as follows: $\hat{C}_{max}$(plains)=8286 cm^{-3} and $\hat{C}_{max}$(hills)=3833 cm^{-3}. Station 7 was visited on the morning of February 11, and Stations 12, 13, and 14 were visited on the morning of February 12; all had low Aitken counts. The Rapid City soundings from those two dates showed no subsidence inversions existing at 0500 MST, but did show inversion development during the day at about 800 mb. It is possible that Stations 12, 13, and 14 were visited before the development of the inversion on February 12 and the remaining stations afterwards, thus explaining the low counts at those three sites. However, this reasoning does not account for the low counts at Station 7 on February 11.

The influence on Aitken counts from the passage of cold fronts in this area is often difficult to quantify because of the fact that precipitation quite often occurs in conjunction with them. Only two cases of "dry" cold frontal passages during sampling runs are evidenced from the data.

Case 1: September 18-19, 1974, sampling run #9. A Canadian cold front had passed Rapid City from the north at approximately midnight on September 19. Respective geometric mean Aitken counts for hills and plains stations on the day before the frontal passage and on the day

afterwards were as follows: $\hat{C}_{max}$(plains)=3023 and 616 cm^{-3}, and $\hat{C}_{max}$ (hills)=1923 and 755 cm^{-3}. One station (S-10) reported fog and was omitted from the analysis.

Case 2: July 30-31, 1975, sampling run #30. A Pacific cold front had passed Rapid City from the northwest at approximately 0500 MST on July 31. Geometric mean Aitken counts for hills and plains stations on the day before the frontal passage and on the day afterwards were as follows: $\hat{C}_{max}$(hills)=2379 and 1223 cm^{-3}, and $\hat{C}_{max}$(plains)=10,660 and 971 cm^{-3}.

In both cases the cleanup was fairly evident, especially for the plains stations. It is reasonable to assume that in this area cold fronts and the associated air masses behind them are reservoirs of clean air. This is apparently true because regions to the north and northwest of Rapid City and the Black Hills are sparsely populated, with no large industrial areas and few sources of fugitive dust. In addition, the passage of cold fronts generally results in a great deal of turbulent mixing and dispersion of whatever particles are present. The fact that the hills counts are slightly higher than the plains counts after frontal passage may be due to the slope of the frontal surface, leaving a smaller vertical mixing depth over the hills than over the plains. One more point worth mentioning is that counts rise again to "normal" levels approximately one to two days following a cold frontal passage as observed from the data.

There may be some argument that since cold frontal passages temporarily decrease Aitken counts, data taken within a certain period of time after such events should be omitted from statistical analyses as were precipitation data. This should not be the case since all types of

synoptic scale weather patterns affect counts to some degree. To throw one type of synoptic condition out and include others would bias the data. As it now stands, the observations represent many different weather types.

3.4 Natural Decrease in Counts with Increasing Elevation

Station geometric mean Aitken counts have been plotted against elevation in Fig. 6. A fairly significant negative correlation coefficient at 99% confidence is evident here, indicating that the decrease associated with increasing elevation is fairly strong and reliable.

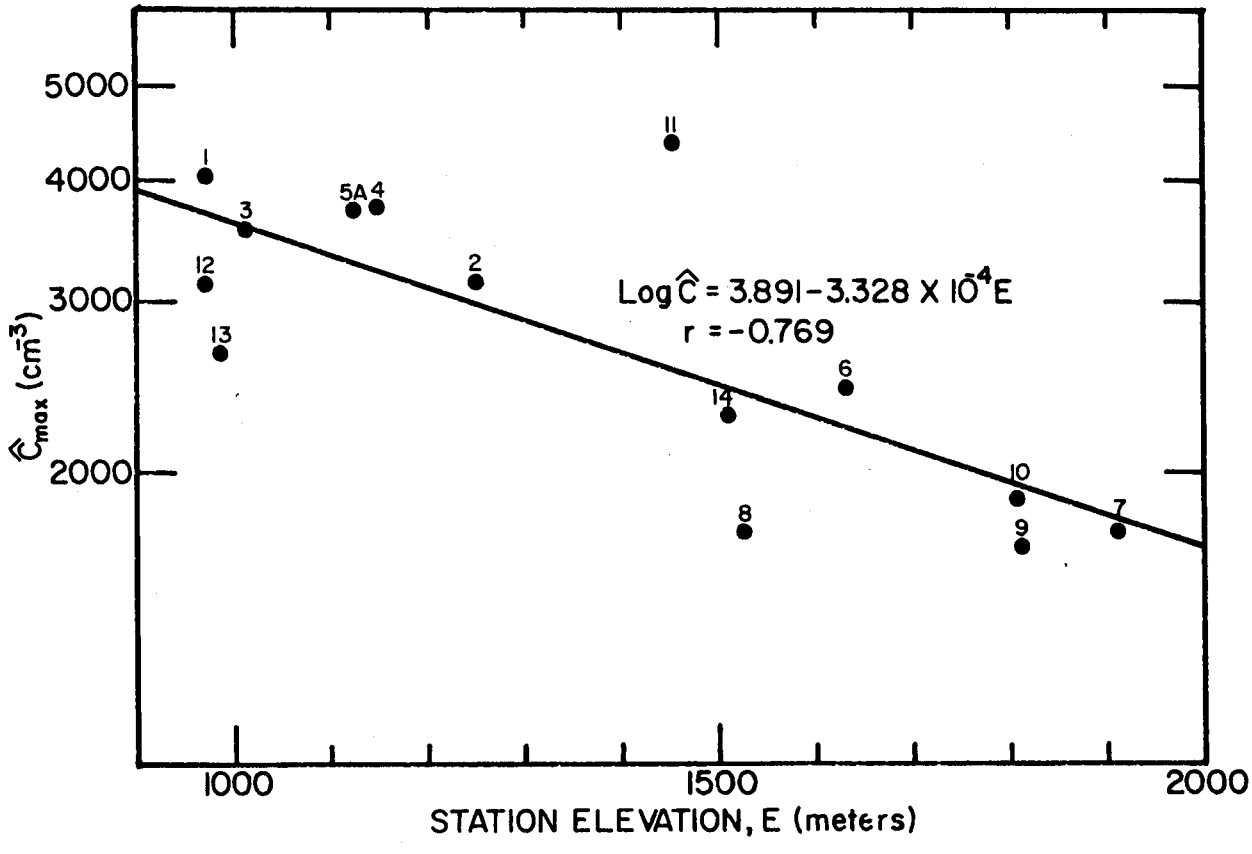


Fig. 6. Geometric mean Aitken nuclei concentration as a function of station elevation.

3.5 Variation Due to Local Meteorological Conditions

Linear regressions were run to determine what, if any, effect various meteorological conditions have had on Aitken counts for the Black Hills data. Table 3 summarizes the results of these regressions.

TABLE 3

Summary of Regression Analyses

Y	X	a_o	a_1	r	s	n	Comments
Log C_{max}	RH	3.887	-0.0062	-0.20	0.48	326	Plains
Log C_{max}	RH	3.885	-0.0104	-0.43	0.39	283	Hills
Log C_{max}	T	3.512	0.0027	0.07	0.49	326	Plains
Log C_{max}	T	3.195	0.0108	0.29	0.41	283	Hills
Log C_{max}	W	3.600	-0.0115	-0.06	0.49	326	Plains
Log C_{max}	W	3.211	0.0492	0.21	0.42	283	Hills
Log C_{max}	Log z	4.358	-0.2860	-0.28	0.65	12	S-1, S-13

For the above regressions, the following conditions apply:

Linear Regression Equation: $Y = a_o + a_1 X$

r = correlation coefficient

s = standard error of estimate

n = number of observations

C_{max} = Aitken nuclei counts

RH = relative humidity (%)

T = temperature (°C)

W = wind speed (m sec^{-1})

z = mixing depth (m)

In the case of the regressions with relative humidity, both the hills and the plains stations demonstrate negative correlation at 99%

confidence. The temperature regressions show a positive correlation at 95% confidence for the hills stations, with no significant correlation apparent for the plains stations. The wind speed regressions show a positive correlation at 99% confidence for the hills stations, with no significant correlation apparent for the plains stations. The mixing depth regression for Stations 1 and 13 shows no significant correlation at 95% confidence. The reasons will be discussed in the next section.

3.6 Variation Due to Mixing Depth

An attempt was made to determine the effect of mixing depth on Aitken counts, since it is a well-known fact that pollutant concentrations show an inverse relationship with mixing depths. Mixing depth is defined as the vertical extent of the atmosphere available for the dispersion of pollutants, and is determined by the height of an inversion layer, whether it be the tropopause, or a subsidence, frontal, or radiation inversion.

The only available data for this study were from radiosonde soundings obtained from the Rapid City weather service office, which conducts daily radiosonde releases at 0500 and 1700 MST. Stations 1 and 13, being within 50 miles of Rapid City on the eastern side of the Black Hills, were the only stations used. Inversion heights within 2000 meters above ground level were determined from the soundings and were compared to Aitken counts at the two stations. There are several possible explanations for the low correlation coefficients obtained, with the most probable one being the lack of plottable data points.

3.7 Variation Due to Wind Direction

A wind rose diagram of wind direction vs. number of observations for the 1974-1976 data is shown in Fig. 7 with an inset showing the

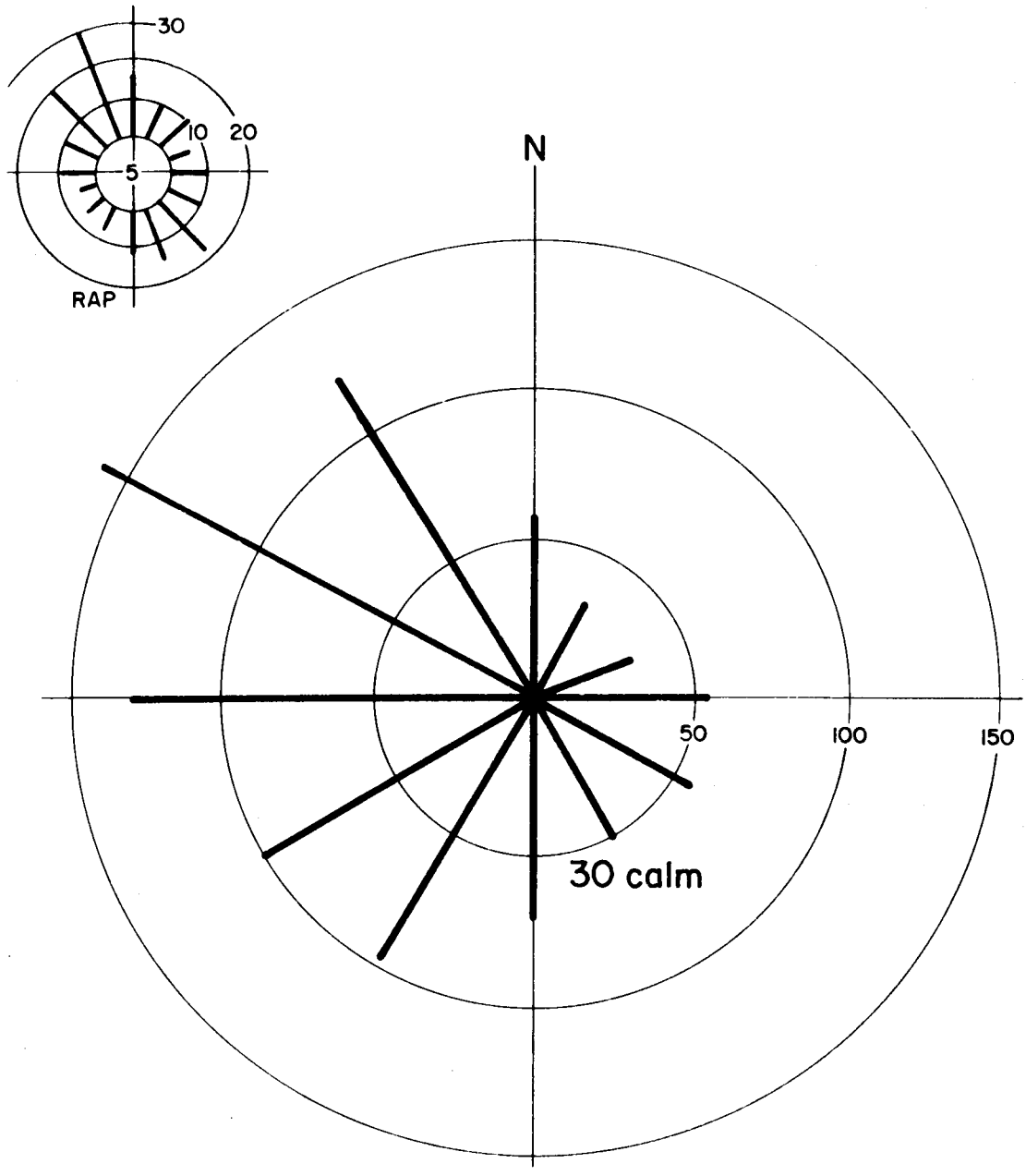


Fig. 7. Wind rose showing frequency of observations as compared to wind direction for all Black Hills stations. Inset shows Rapid City wind rose for 1951-1960 with figures representing percentages of total observations.

normal Rapid City wind rose for 1951-1960. The major difference between the two is a shift of winds from the southeast (moisture source) to the southwest (moisture sink) in the two year rose. This primarily occurred during the first year of sampling, when a minor drought was experienced in the area during the summer months (1974). When geometric mean Aitken counts are compared for each of the two sampling years, as in Table 4 below, the results of the dry first year are evidenced by the higher mean counts. In this case, dusts from wind erosion were the major contributing factor to the higher counts.

TABLE 4

Yearly Geometric Means (All Stations)

	Hills	Plains
First Year	2001 cm^{-3}	3621 cm^{-3}
Second Year	1583 cm^{-3}	2944 cm^{-3}
Overall	1758 cm^{-3}	3252 cm^{-3}

Figure 8 represents a plot of Aitken and near-CCN counts against wind direction for the hills and plains stations. This figure suggests that the "dirtiest" winds are those from the south or southwest.

3.8 Diurnal Variation

In order to test the variation of Aitken counts during the day, the first and last stations of each sampling circuit (S-1, S-7, S-8, S-13) were compared as to the difference in counts in the morning hours vs. the afternoon hours, depending on whether the stations were visited on a normal circuit or a reverse circuit. The results are presented in Table 5. The counts averaged nearly 2 1/2 times higher in the afternoon hours than in the morning hours.

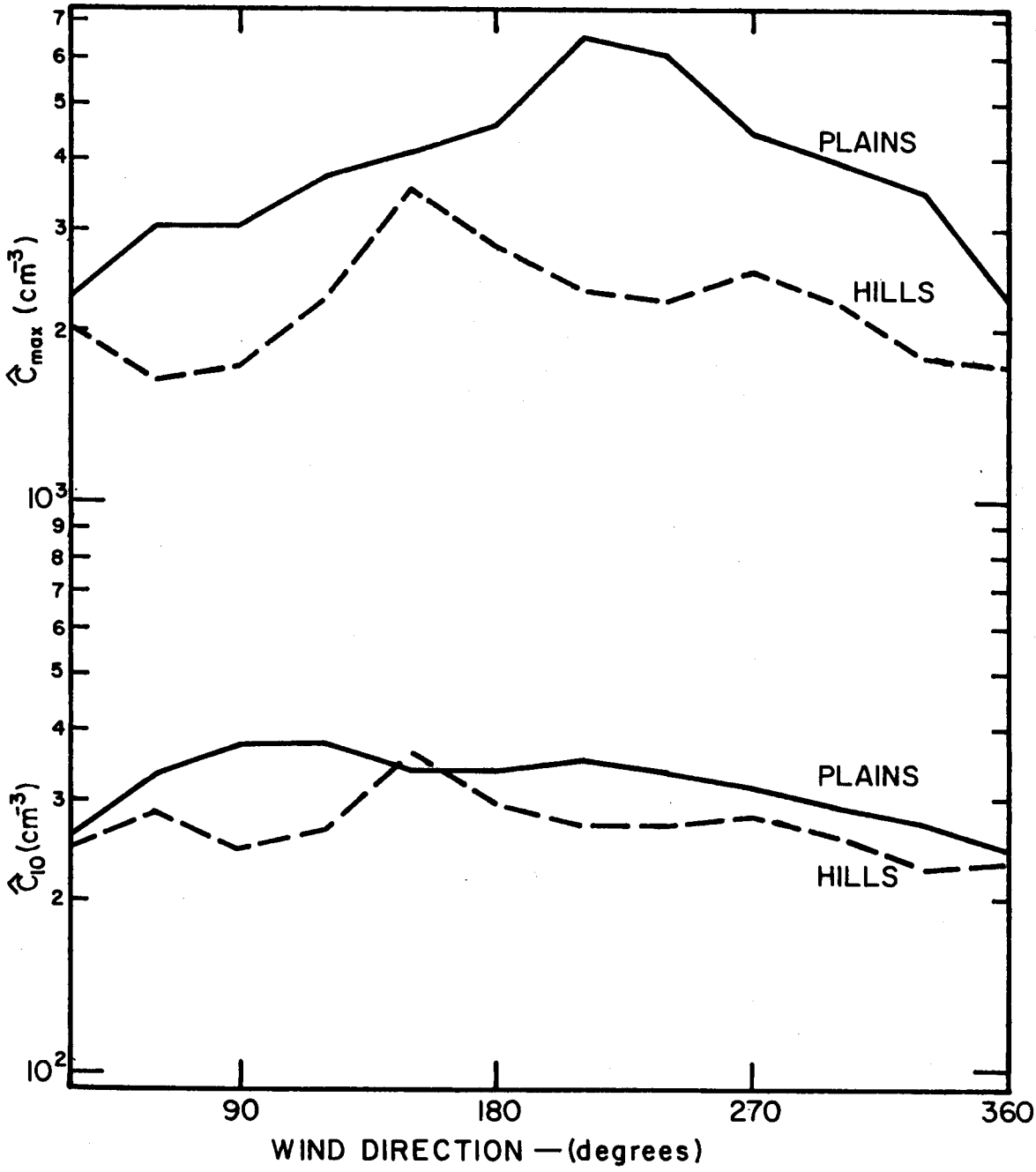


Fig. 8. Geometric mean Aitken ($\hat{C}_{max}$) and near-CCN ($\hat{C}_{10}$) nuclei concentrations as a function of wind direction stratified by plains and hills stations, plotted by 60° intervals with 30° moving averages.

TABLE 5

Diurnal Variation Test

	morning		afternoon	
Station	Time	$\hat{C}_{max}$	Time	$\hat{C}_{max}$
1	08:53	3870	15:00	4737
7	09:12	1093	14:49	3269
8	08:33	1164	15:03	3441
13	08:49	1706	14:44	4522

4. GREEN AREA TESTS

4.1 General Explanation

As previously stated, the primary purpose of this study is to determine whether or not the "green area" scavenging effect is evident in the Black Hills forested region. In order to fulfill this objective, several tests have been devised, most of which were contained in IAS Technical Report 75-8, with the addition of one new test and a more rigorous treatment of the original ones. In all the tests, precipitation scavenging events were omitted, and elevation corrections were made where applicable.

4.2 Particle Shadow Test

This test involves one or more stations along the periphery of the Black Hills. Aitken nuclei concentration is plotted against wind direction for stations at the northern and southern ends of the hills. Should a green area effect exist, these data would show that the highest counts occur with winds moving from the plains onto the station, with decreased counts expected from the forested areas. Since the elevation at the stations under consideration is relatively constant, and since precipitation scavenging events have been omitted, any noticeable filtering effect from the forested areas should be due to the green area scavenging process.

The idealized situation is illustrated in Fig. 9, showing a concentration minimum at 360° for the southern plains stations, and a minimum at 180° for the northern plains stations. In the actual test, Stations 3 and 4 and Stations 12 and 13 were combined. The results are shown in Figs. 10a and 10b. No conclusive particle shadow is evident

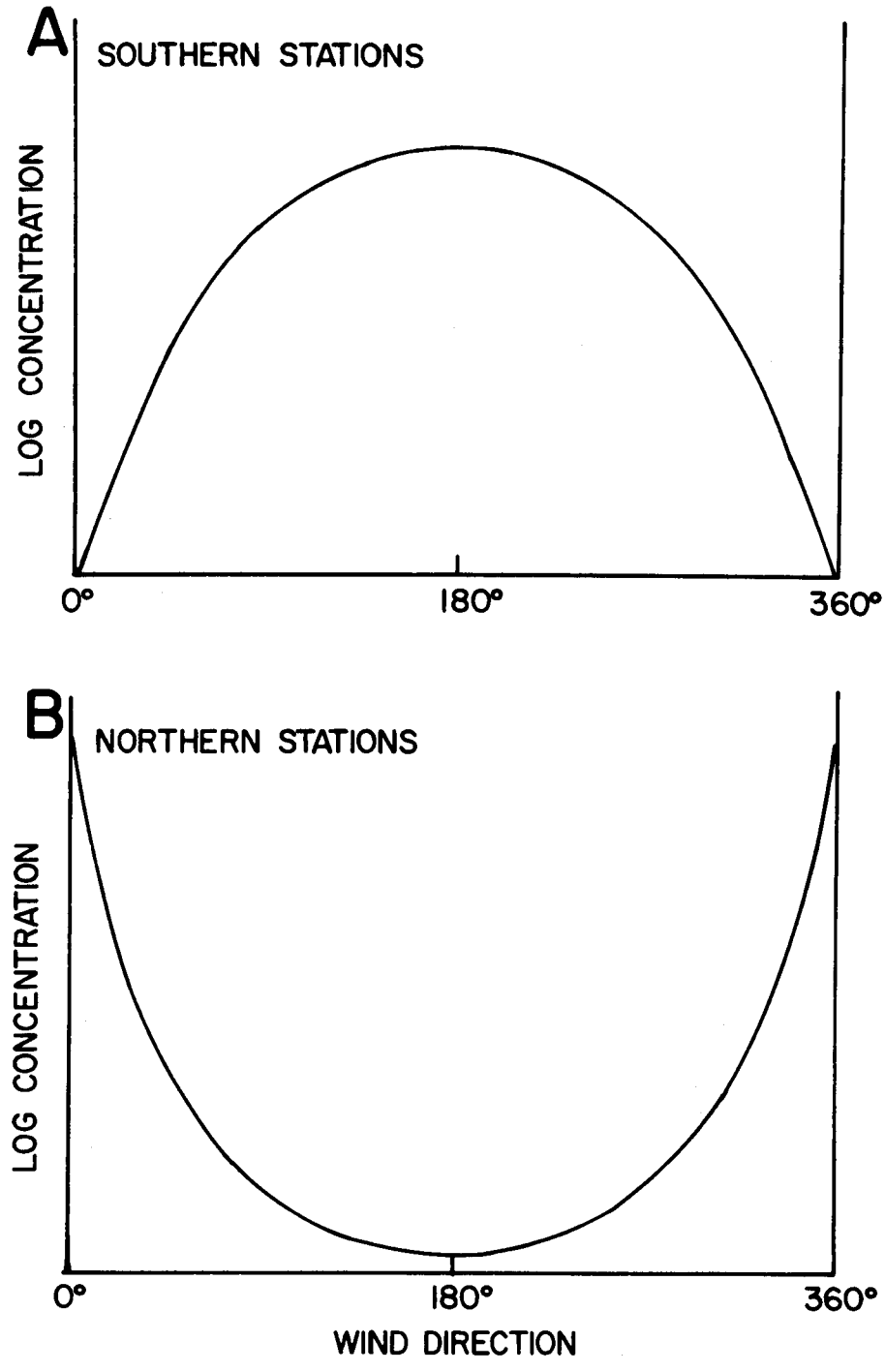


Fig. 9. Idealized distribution of Aitken nuclei concentration versus wind direction for combined southern and northern plains stations in particle shadow test.

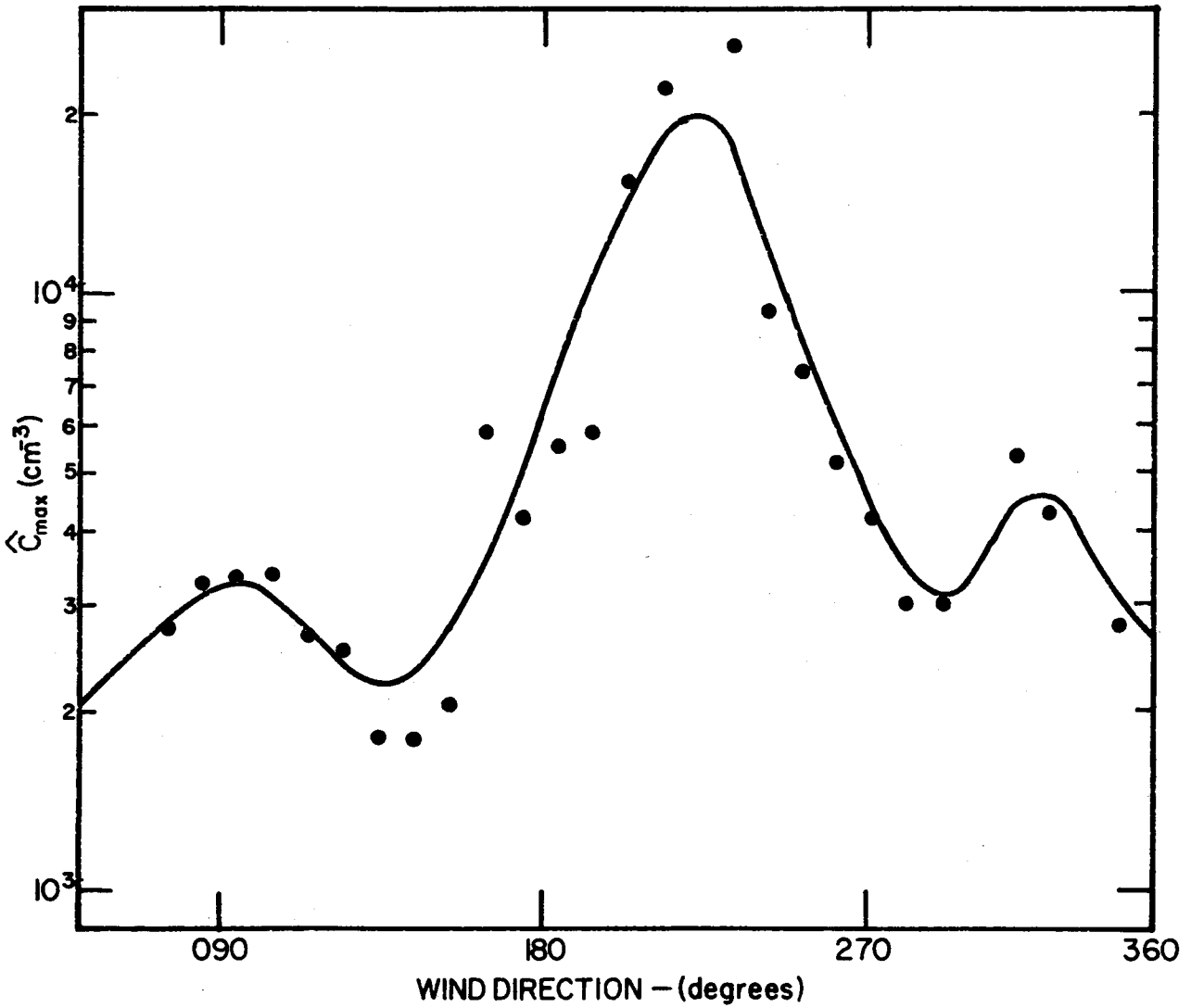


Fig. 10a. Geometric mean Aitken nuclei concentration versus wind direction for the particle shadow test using Stations 3 and 4. Plotted by 30° intervals with 10° moving averages. No data from 360° to 070°. Curve fitted by eye.

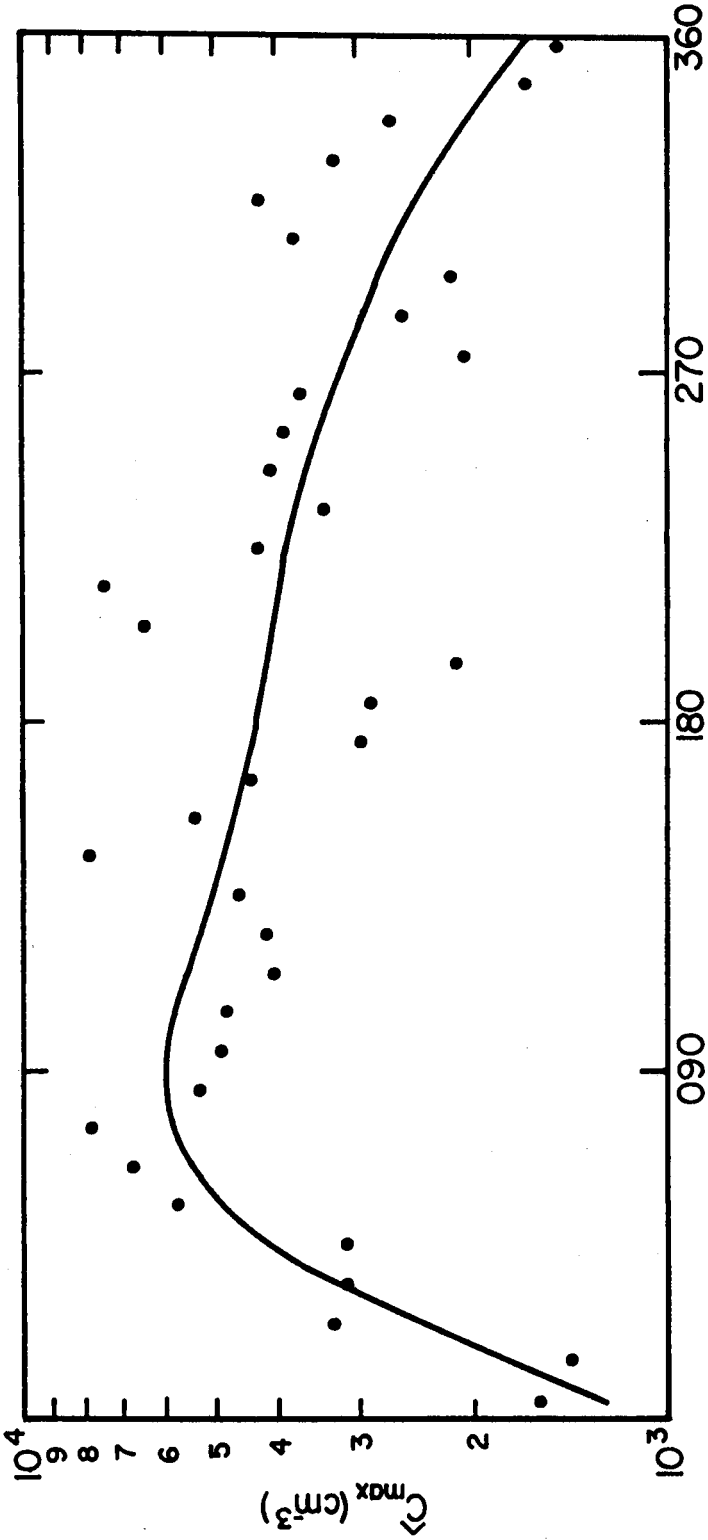


Fig. 10b. Geometric mean Aitken nuclei concentration versus wind direction for the particle shadow test using Stations 12 and 13. Plotted by 30° intervals with 10° moving averages. Curve fitted by eye.

for Stations 12 and 13. This unexpected result is apparently due to existing local pollution sources between the green area and the stations. Stations 3 and 4 show a pronounced particle shadow for northerly winds as expected.

A statistical test was applied to the data in Fig. 10a to determine whether or not the points in the interval 180-250° were from the same population as the remaining points. The null hypothesis was that they were from the same population. A z-statistic was used after normality of the distribution was established. H_o was rejected at $\alpha = .00005$.

4.3 Opposing Wind Sector Test

This test is similar to the particle shadow test except that in this case two pre-defined wind sectors for each station are used rather than the entire compass. Aitken concentration is plotted against wind direction only in the chosen sectors; one sector from the green area and one sector from the plains. All possible local sources of pollution are excluded in selection of the sectors. The geometric mean for each sector is calculated and then compared to that of its opposing sector by use of the Wilcoxon-Mann-Whitney rank-sum test, to determine significance levels at which the null hypothesis can be rejected. In this case, the null hypothesis is that the data points in both sectors are from the same population. The basic procedure in the rank-sum test is that all the observations from both sectors are ranked, then the sum of the ranks of the smaller sector is calculated and compared to tabulated significance levels to determine rejection criteria.

The opposing wind sector test was applied to Stations 1, 2, 3, 5A, 11, 12, and 13, all located on the periphery of, or slightly into the green area. The results of these tests are shown in Fig. 11 and the significance levels from the rank-sum tests are presented in Table 6.

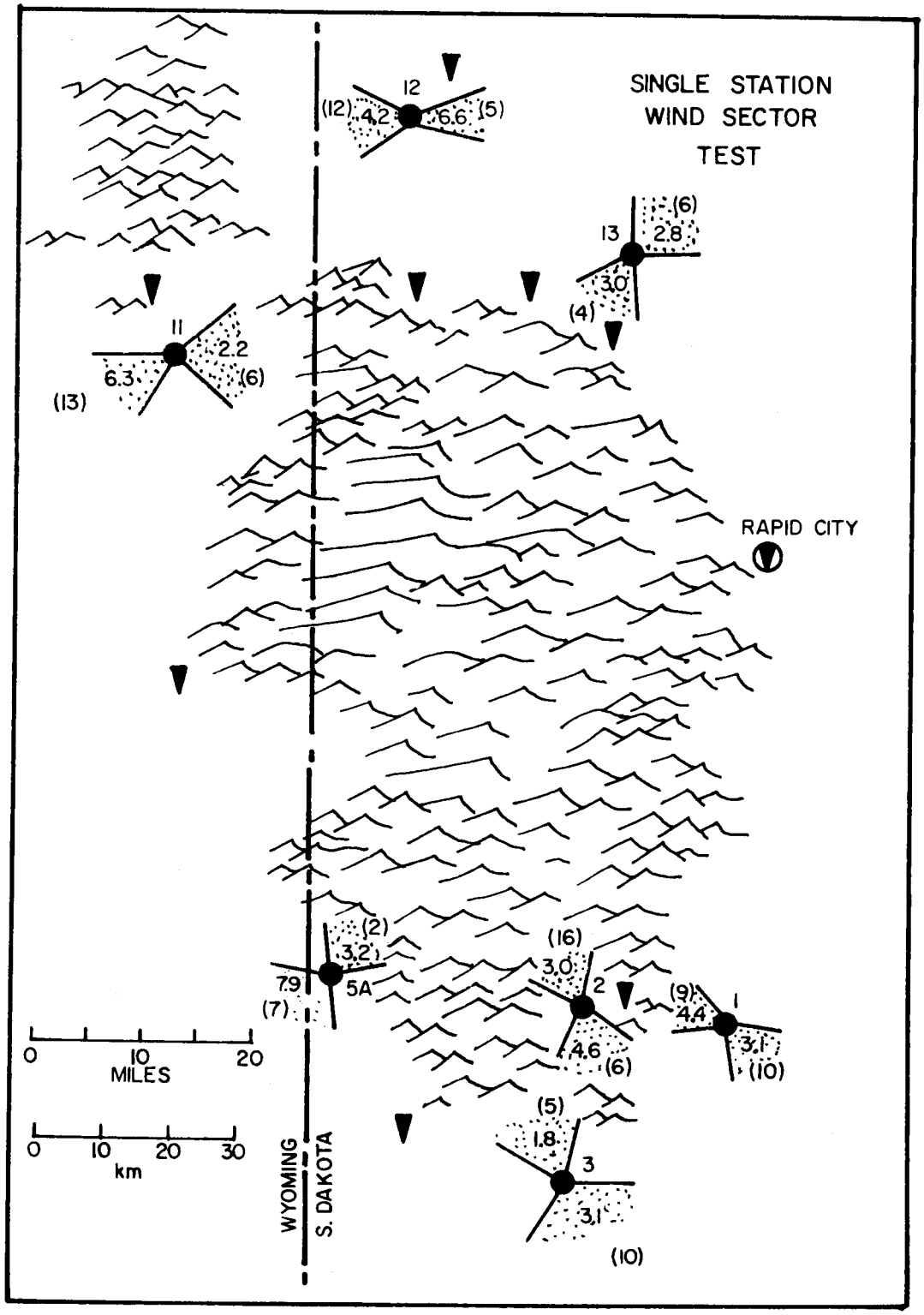


Fig. 11. Results of opposing wind sector tests for Stations 1, 2, 3, 5A, 11, 12, and 13. Sample size given in parentheses near each wind sector, and geometric mean Aitken nuclei concentration in units of 10^3 cm^{-3} given within wind sector.

TABLE 6

Significance Levels from Rank-Sum Tests

Station	α
1*	.16
2	.06
3	.34
5A	.17
11	.02
12	.11
13*	.46

(* indicates a negative effect)

Station 4 was omitted from these tests because no data points were found in the green area sector, which has been re-defined for the total 24-month data. Stations 3 and 5A show positive results, but with higher values of α, which is due to the limited sample size in one or more of the wind sectors. Stations 1 and 13 indicate a negative effect which is thought to be due to the presence of local pollution sources between the green area and the sampling sites. In both cases the sources could not be separated from the remaining data. The remaining three stations (Stations 2, 11, 12) show a significant (0.11 or better) green area effect.

4.4 Test Using Elevation Correction

In this test, the geometric mean of Aitken counts at a hills station is compared to that at one or more plains stations. But, since the hills stations are higher in elevation, a correction must be applied because of the natural decrease in nuclei with increasing elevation.

The correction factor obtained in the Bear Butte traverse (Davis et al., 1975) is used here, which has the effect of placing all the stations "at the same elevation." The Bear Butte elevation data, while not the only data available on nuclei decrease with height, is the only known source of information on this effect in this region of the country, thus it was used in preference to other sources.

Stations 3, 4, 5A, and 11 are the four plains stations farthest from any local pollution sources, and thus they were the ones chosen for this test. They were compared to Station 9, the most isolated of the green area stations. After applying a percentage elevation correction from Bear Butte, the results are as listed in Table 7. This test demonstrates that a green area cleanup is present, since the counts at the four plains stations after the correction remained higher than the counts at Station 9.

TABLE 7

Test Using Elevation Correction

Station	Actual Count	Elevation (m)	Corrected Count
9*	1687	1817	1687
3	3583	1007	1737
4	3766	1142	2150
5A	3727	1125	2087
11	4381	1458	3382

(* green area station)

4.5 The Use of Rapid City Stations for Green Area Tests

Three stations in the Rapid City area, which have been involved in an air quality study by the IAS for the State of South Dakota, were used in these tests. Figure 12 shows the locations of these three

stations just outside the northwest portion of Rapid City. Station 7x is located in an area open to the west and south and has characteristics similar to a plains station, Station 6x is located just inside the green area, and Station 5x is located deeper into the green area.

4.5.1 Rapid City Plume Cleanup Test

In this test wind sectors are defined for each of the three stations to deliberately include the western Rapid City plume, in order to determine if there is a cleanup effect as the plume travels deeper into the green area. Stations 6x and 7x are at the same elevation, but Stations 5x is 130 meters higher, so an elevation correction was applied as was done in the previous section. The results, listed in Table 8, do indeed show an increasing cleanup of the Rapid City plume as it progresses farther into the green area.

TABLE 8

Rapid City Plume Cleanup

Station	Wind Sector	$\hat{C}_{max}$	n
5x	030-150°	6399	10
6x	060-180°	9427	13
7x	180-270°	13640	14

4.5.2 Equal Elevation Test

This test is quite similar to the opposing wind sector test described in Section 4.3. In this case, a green area sector was defined for Station 6x and a plains sector was defined for Station 7x. The geometric mean Aitken counts from each sector were compared directly, since both stations are at the same elevation. The results listed in Table 9, show that the green area brings the cleanest air to the stations.

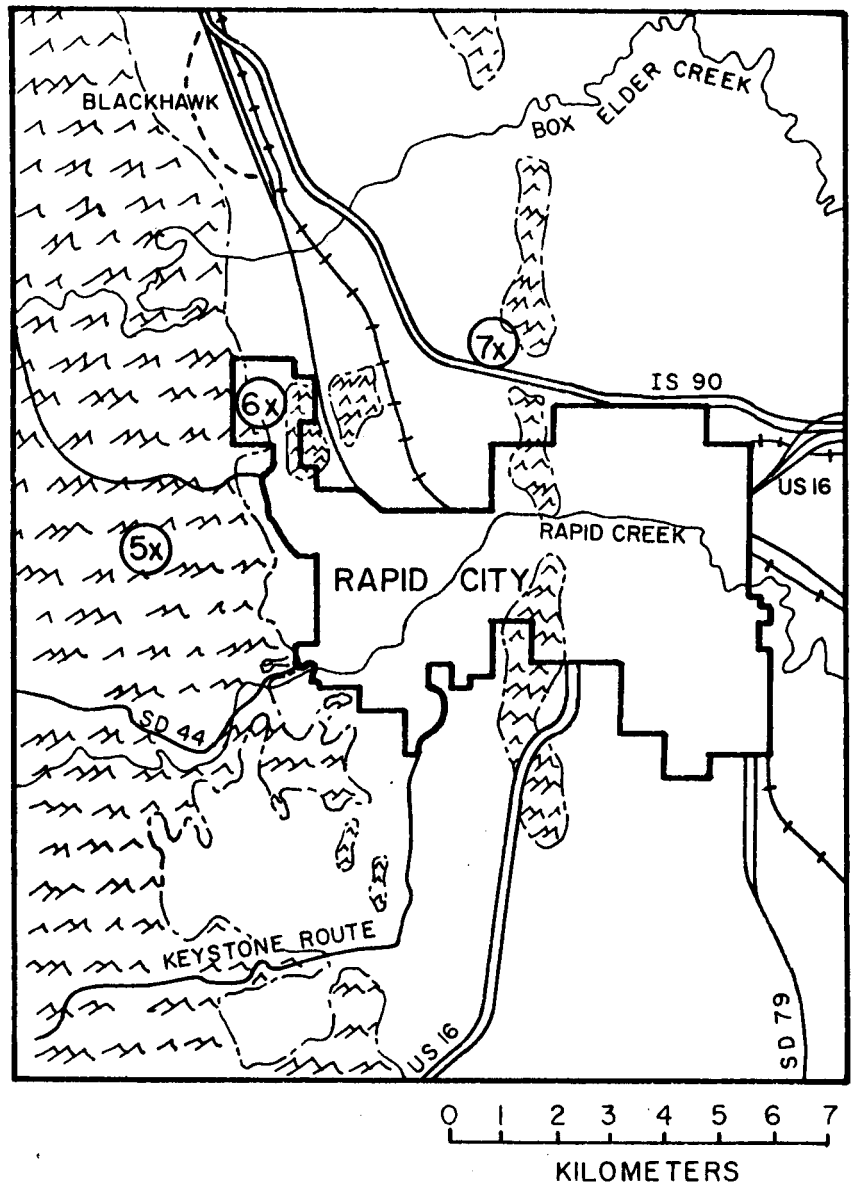


Fig. 12. Locations of three Rapid City air quality sites (Stations 5x, 6x, and 7x) from the state project. All three are approximately the same distance from the center of northwest Rapid City.

TABLE 9

Equal Elevation Test

Station	Wind Sector	$\hat{C}_{max}$	n
6x	210-330°	3025	35
7x	315-045°	6116	32

5. SUMMARY AND CONCLUSIONS

Analysis of the Black Hills data has shown that aerosol concentrations are affected by a number of variables. Precipitation events have shown strong scavenging effects, with snow being the most effective when compared to liquid forms. A natural decrease in nuclei with increasing elevation was also noted. Other meteorological variables, such as wind speed, temperature, relative humidity, and mixing depth have shown weak to no correlation (significant at $\geq$95% confidence) with nuclei concentration. However, variations in counts were indicated in case studies of certain synoptic scale weather phenomena, such as decreased counts following cold frontal passages and increased counts when Great Basin highs were present.

After correcting for precipitation scavenging and the natural decrease in nuclei with increasing elevation, five tests were performed on the data in an attempt to either support or reject the hypothesis that the Black Hills forests act as a "green area" sink for Aitken nuclei. Of all the tests run, the few that demonstrated a negative green area effect can be discarded because of contamination influences from local sources. The particle shadow test for the southern stations showed a positive particle shadow, but the northern stations showed inconclusive results. The opposing wind sector tests showed three stations indicating a strong green area effect, two with positive but statistically weak results due to small sample size, and two with negative (explainable) results.

The remaining three tests also support the green area effect. The test using elevation correction showed an average green area cleanup of 23%. The Rapid City plume cleanup test suggested a 50% reduction in

nuclei counts between the plains station and the deepest green area station. The equal elevation test from the Rapid City data also indicated a 50% particle cleanup by the green area.

Overall, concentrations in the Black Hills compare favorably with those in other forested, mountainous areas. Tester (1964) observed Aitken concentrations in the Adirondack Mountains to average between 2500 and 4100 cm^{-3} in a study done in July 1963. Hogan (1966) observed median Aitken counts in Yellowstone Park to be near 2000 cm^{-3} in the summertime and approximately 600 cm^{-3} in winter. In this current study, Station 9 has shown a summertime average of 2580 cm^{-3} and a wintertime average of 1367 cm^{-3}, which shows the higher Black Hills to be fairly clean when compared to the above studies.

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APPENDIX A

PARTICULATES IN THE BLACK HILLS REGIONS

General Sources

The Environmental Protection Agency has provided a brief table of particulate sources for Pennington County, South Dakota of which Rapid City is the county seat. This information is presented below in Table 1. Since Rapid City is included in this inventory, the tonnage totals will be higher than for the Black Hills region in general, but the sources will be similar.

TABLE 1

Particulate Sources in Pennington County
(Contributions in 1974)

A.	Area Sources	
	1. Fuel Combustion	46 tons
	2. Mobile Sources (Transportation)	.368 tons
	3. Combustion of Solid Waste	.113 tons
	4. Fugitive Dust	27,217 tons
	Total	27,744 tons
B.	Point Sources	
	1. Fuel Burning	.449 tons
	2. Process Losses	5055 tons
	Total	5504 tons

Terpenes as a Source of Particulates

It is an accepted fact that plants release appreciable quantities of volatile organic substances to the atmosphere (Went, 1966). A large percentage of these substances fall into the general category of terpenes, and more specifically: α and β-pinene, isoprene, and limonene. Went

proposed that terpene emissions can react with atmospheric constituents in the presence of sunlight in a process similar to photochemical smog production initiated by hydrocarbons from automobile exhaust. Aitken nuclei are assumed to be by-products of this process and consequently are believed to be responsible for the "blue haze" phenomenon that often appears over forested regions. In fact, the "smoke" in the Great Smoky Mountains of North Carolina is primarily due to this process.

The primary source of terpenes emitted from trees is believed to be the foliage. These emissions occur in conjunction with the plant's normal photosynthetic and respiratory processes. Some terpenes, like α-pinene, are emitted from foliage in the dark, while others are not (Rasmussen, 1972).

In the forested Black Hills area, α-pinene is the most common terpene type, being emitted by the following species: ponderosa pine, juniper, and spruce, which together comprise over 90% of the local forests. Aspen and oak are isoprene emitters, but account for a very small fraction of the local forests.

Rasmussen (1972) conducted experiments to determine emission rates of α-pinene using uncrushed foliage in a closed chamber with an artificial light source, constant humidity of 92%, and varying temperatures. He found that higher temperatures resulted in greater emissions, which he attributed to increased plant transpiration and metabolism and the higher vapor pressure of α-pinene at the increased temperatures.

During the summer of 1975, the author conducted a number of experiment in the Cloud Physics Laboratory of the IAS to measure terpene-produced Aitken particles under varying conditions of temperature and humidity. Distilled α-pinene was used in these experiments since it is the

predominant terpene in the forests of this area. Table 2 lists some of the properties of α-pinene.

TABLE 2

Physical and Chemical Properties of α-pinene

1. Chemical formula: $C_{10}H_{16}$
2. Molecular weight: 136.24 gm mol^{-1}
3. Melting point: -55°C
4. Boiling point: 156.2°C
5. Density: .8582 gm ml^{-1} (at 20°C)
6. Vapor pressure: 1013.25 mb (at 156.2°C)
   133.33 mb (at 90.1°C)
   13.33 mb (at 37.3°C)
   1.33 mb (at -1.0°C)

In addition to the aforementioned properties, α-pinene has a few miscellaneous chemical properties as follows: it readily undergoes oxidation, forming a peroxide and oxidation products which are responsible for its odor. In moist air and sunlight it forms pinol hydrate ($C_{10}H_{16}(OH)_2$).

The following equipment was used to conduct the terpene lab experiments:

(1) A test chamber of .142 cubic meters volume equipped with a thermometer, a ventilation system, and heating coils.

(2) A fogging system comprised of a distilled water reservoir and a vacuum pump to force air to mix with water from the reservoir to form a "jet" of finely dispersed water droplets.

(3) A Gardner counter to measure air samples from the chamber for Aitken particles.

(4) A dew-point hygrometer to determine the humidity in the chamber.

(5) A small circulating pump to draw air samples from the chamber.

Figure 1 illustrates the arrangement of equipment used in these experiments.

The experimental procedures used in these experiments were as follows:

(1) The chamber was flushed with dry nitrogen.

(2) Readings were taken with the Gardner counter to determine that Aitken counts were low.

(3) A prescribed amount of α-pinene was added to the chamber through a small opening in the top.

(4) The ventilation system inside the chamber was turned on to circulate the air and allow adequate evaporation of the α-pinene.

(5) Readings were again taken with the Gardner counter.

(6) Humidity was varied utilizing the water reservoir-vacuum pump system, and temperature was raised by means of the heating coils in the chamber.

(7) Readings were again taken with the Gardner counter.

After running the experiment as outlined in procedures 1-7 many times, there appeared to be a strong positive correlation between Aitken counts and relative humidity, and an even stronger correlation between Aitken counts and temperature. However, after conducting further tests, it was determined that the heating coils in the chamber were adding artificial nuclei when turned on. Heated surfaces do produce ions and promote reactions in the air, leading to an increase in nuclei (Landsberg, 1938). Thus, all data collected after the addition of artificial heat was discarded as erroneous. Most of the remaining data were considered to be fairly reliable and were subsequently analyzed.

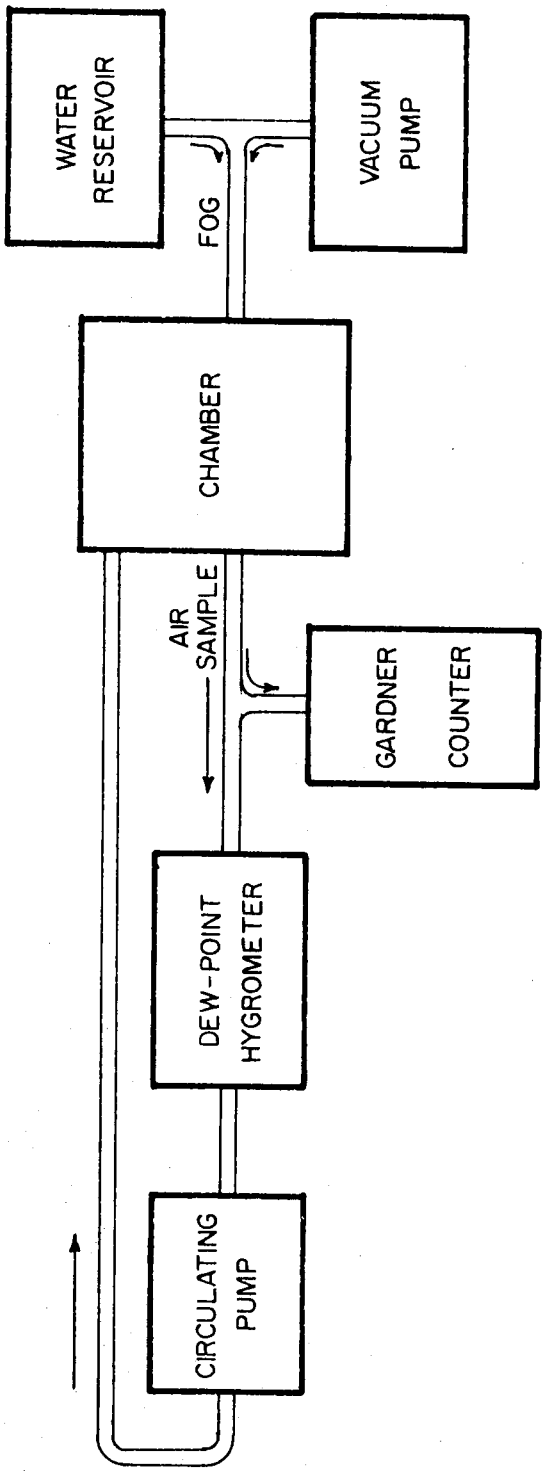

Fig. 1. Laboratory equipment arrangement used in terpene experiments.

Table 3 contains the results of the statistical regressions performed following the omission of the erroneous data. The correlation coefficients, when compared to tabulated maximum values obtained randomly, show that the correlation is not significant at 95% confidence for either the Aitken count (log C) versus temperature (T) regression or the Aitken count (log C) versus relative humidity (RH) regression. However, a 0.54 correlation at 95% confidence is evidenced when the three variables are considered collectively.

TABLE 3

Summary of Regression Analyses from Terpene Experiments

Linear Regressions

Y	X	a_o	a_1	r	s	n
log C	T(°C)	2.213	0.0093	0.12	0.41	38
log C	RH(%)	1.845	0.0123	0.32	0.39	38

Multiple Linear Regression

Z	Y	X	a_o	a_1	a_2	r	s	n
log C	RH(%)	T(°C)	0.2180	0.0385	0.0288	0.54	0.13	38

In the above regression analyses the following conditions are true:

Linear Regression Equation: $Y = a_o + a_1X$

Multiple Linear Regression Equation: $Z = a_o + a_1X + a_2Y$

r = correlation coefficient

s = standard error of estimate

n = sample size

These laboratory experiments have shown that there is at least a small positive correlation between aerosol formation from α-pinene vapor and temperature and relative humidity when combined. Therefore, it can

be expected that the summer months with their potential for a combination of warm temperatures and high daytime humidities, will result in a greater generation of Aitken aerosols from the α-pinene vapor present in the forests of the Black Hills, than during other seasons. This is supported by the fact that the strong aroma of pine, which is related to the presence of terpenes, is most prevalent during the summer months.

APPENDIX B

DEVELOPMENT OF A LOADING INDEX

Form of the Index and Explanation of Terms

The development of a method of estimating particulate concentration given certain meteorological observations has certain advantages. A rough, empirical particulate loading index was defined for the Black Hills data as follows (Davis et al., 1975):

$$I = \frac{K\ P(1+C)}{(1+S)\ \exp(f(T,W))}$$

K represents a constant of proportionality, C is observed fractional cloud cover, S is observed fractional snow cover, T is temperature in °C, W is wind speed in meters sec^{-1}, and P is a precipitation scavenging factor defined as follows: P=1 for no precipitation, P=2 for light precipitation, P=4 for moderate precipitation or fog, P=10 for heavy precipitation. The S term is included due to the observed phenomenon of snow sublimation releasing particles to the atmosphere. The exponential term was derived from the regression equations of Log C_{max} vs. temperature and wind speed.

Results of Regression Analyses

The respective loading index equations for the hills and plains are as follows:

$$\bar{I}_H = \frac{5x10^6\ P(1+C)}{(1+S)\ \exp(14.75+.113W+.025T)}$$

$$\bar{I}_P = \frac{5x10^6\ P(1+C)}{(1+S)\ \exp(16.38-.028W+.007T)}$$

In these two equations, the mean values of each variables are used, since the index is being calculated for the hills and plains stations together rather than individual sites.

The loading indices for the hills and plains stations as well as Stations 9 and 12 were calculated for each sampling period and correlated against Aitken nuclei counts from the same observation. The results of the regression analyses are given below in Table 1.

TABLE 1

Loading Index Regressions

Y	X	a_o	a_1	r	s	n
Log $\hat{C}_{max}$	Log $\bar{I}_H$	3.375	-0.653	-0.74	0.18	50
Log $\hat{C}_{max}$	Log $\bar{I}_P$	3.178	-1.080	-0.59	0.31	50
Log C_{max}	Log I_9	3.430	-0.873	-0.72	0.30	50
Log C_{max}	Log I_{12}	3.214	-1.264	-0.57	0.43	50

Test for Annual Cyclic Variation

A threshold level was chosen for each loading index to allow a separation of wet and dry days. The data points in each category were then fitted to a polynominal of the type:

$$\text{Log } \hat{C}_{max} = a + bx + cx^2 + \frac{d}{x} + \frac{e}{x^2}$$

by use of a multiple linear regression program, to determine whether an annual cyclic trend could be established in the data. The results are shown in Figs. 1 and 2. No definitive trend is noticeable at this time which is probably due to the lack of sufficient data. It is likely that it would take at least two to three additional years of data collection in order to make any conclusions about annual scavenging cycles in the loading indices.

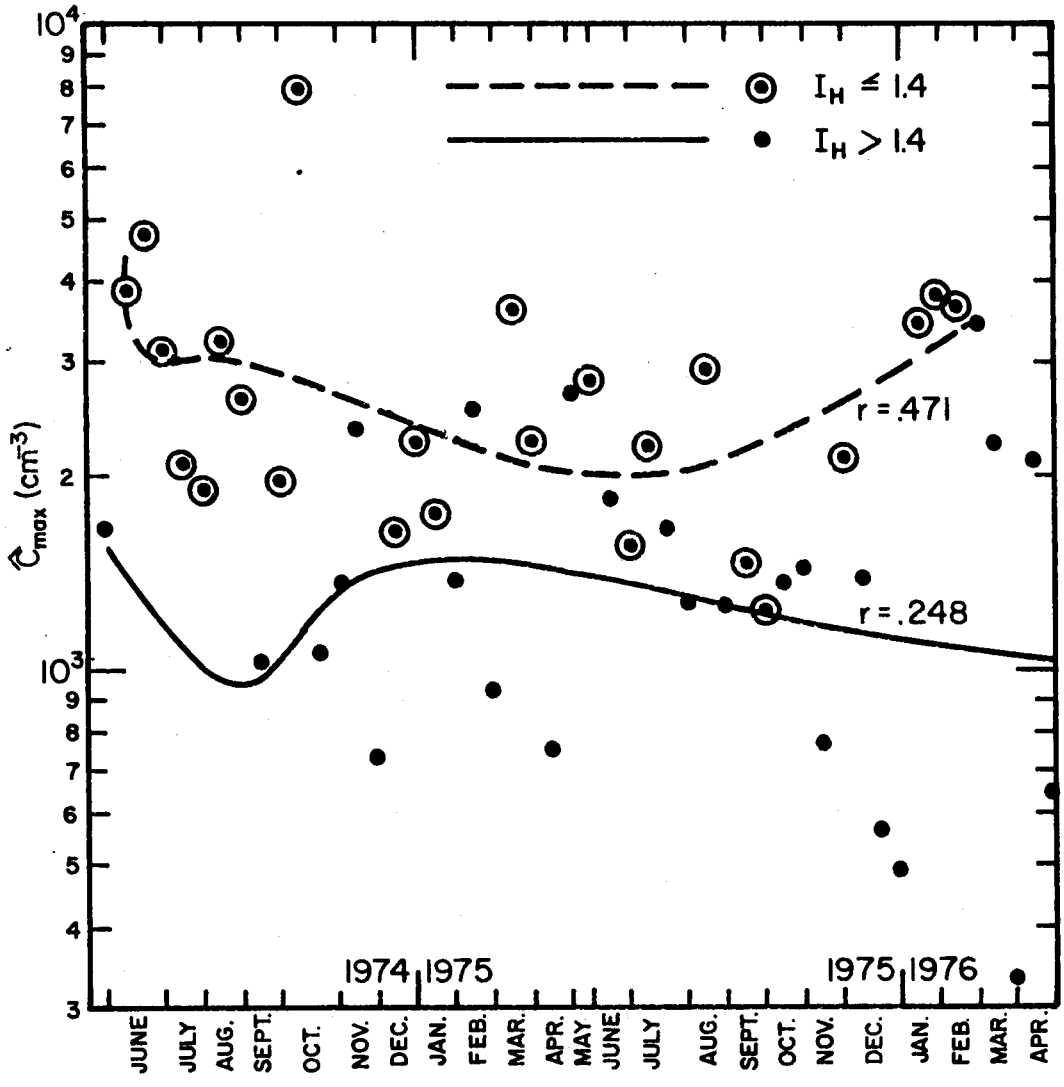

Fig. 1. Geometric mean Aitken nuclei concentration as a function of time through the two sampling year for hills stations, stratified by loading index. Curves represent best polynominal fit.

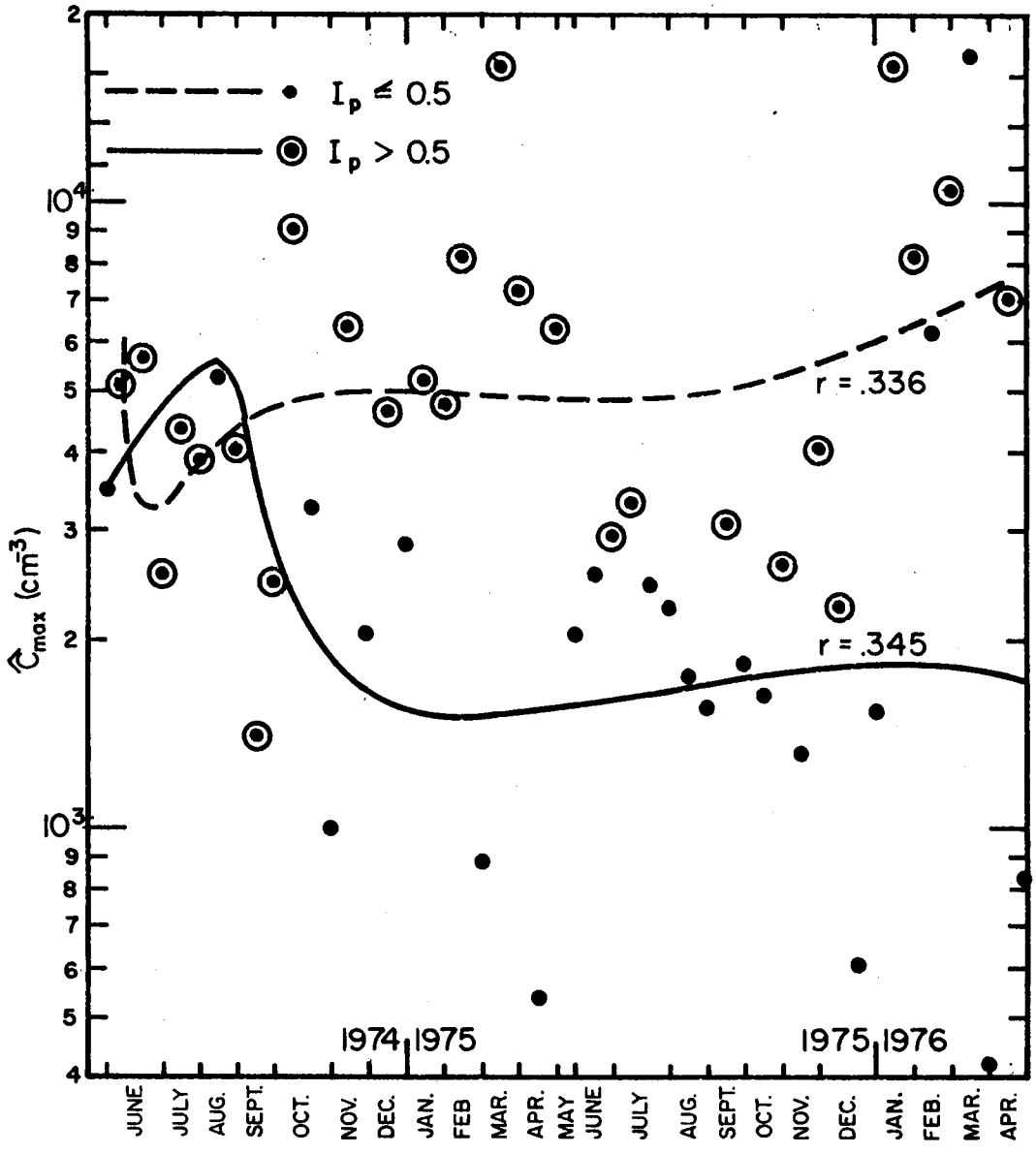

Fig. 2. Geometric mean Aitken nuclei concentration as a function of time through the two sampling years for plains stations, stratified by loading index. Curves represent best polynominal fit.